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13. SUPPLEMENTARY NOTES

14. ABSTRACT

A fast-switching and scattering-free phase modulator using polymer network liquid crystal (PNLC) is demonstrated at λ=1.55 µm for laser beam steering application. The strong polymer network anchoring greatly reduces the visco-elastic coefficient of the liquid crystal. As a result, the PNLC response time is improved by ~300X, although the threshold voltage is increased by ~25X. Computer simulation results agree with experiments well.

15. SUBJECT TERMS

fast-switching, scattering-free, phase modulator, polymer network liquid crystal

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a. REPORT	b. ABSTRACT	c. THIS PAGE			Dr. Shin-Tson Wu
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AFOSR Final Report

HIGH BIREFRINGENCE LIQUID CRYSTALS FOR
LASER HARDENING AND IR COUNTERMEASURE

Contract No.   F49620-01-1-0377
                            UCF Project No. 6501-6057

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Contract Period: July 15, 2001 to August 31, 2004

High Birefringence Liquid Crystals for Laser Hardening and IR Countermeasure

1. Objectives:
The main objective of this program is to develop high birefringence liquid crystal (LC) materials and fast-response devices for laser hardening and infrared countermeasure.

2. Status of Effort:
We have developed a new polymer network liquid crystal for achieving fast-response time for laser hardening and countermeasure.

3. Accomplishments
3.1 Motivation
For laser beam steering using optical phased arrays, a fast-response liquid crystal is needed. We have developed a polymer network liquid crystal (PNLC) which exhibits a fast response time at room temperature. As compared to the conventional approach, our PNLC response time is improved by ~250X. The tradeoff is the requirement of a higher operating voltage.

3.2 Polymer Network Liquid Crystal (PNLC)
Liquid crystal/polymer composites could appear in network or droplet form depending on what polymer and concentration is employed. Liquid crystal droplets could be found when the polymer concentration is higher than 30 wt%. In the lower concentration regime (<15 wt%), the polymer exists as chain-like networks. The polymer field improves the response time; however, the associated light scattering is quite strong in the visible region. Such devices exhibit an anisotropic light scattering behavior and have potential for reflective display and telecom applications. Recently, a stressed LC cell comprising of 14 wt% Nolan-65 polymer was demonstrated to exhibit a fast response time. In this approach, no surface alignment is required. However, a delicate shearing process is needed in order to suppress light scattering. The operating voltage for a 20 μm E7 cell is ~135 V_{rms}.

We have developed a fast-response homogeneously-aligned polymer network liquid crystal (PNLC) cell comprising of ~10% polymers. A unique feature of this PNLC is that light scattering at λ=1.55 μm is suppressed completely. The measured response time is less than 2 ms for a 2π phase change at λ=1.55 μm and T~21 °C. The tradeoff of the PNLC cell is its high operating voltage. To reduce operating voltage, high birefringence and large dielectric anisotropy LC can be considered.

3.2.1 Experimental
To fabricate the PNLC cells, we first mixed a few percent of photocurable monomer to a nematic LC host. The LC/monomer composite was sandwitched between two glass substrates. The inner surfaces of the indium-tin-oxide (ITO) glass substrates were over-coated with a thin polyimide layer and buffed in anti-parallel directions for achieving homogeneous alignment. The filled cell was then exposed to UV light for curing the polymer networks. To eliminate light scattering, we need to control the domain sizes to be smaller than the wavelength. A weak UV intensity would result in coarser polymer networks which, in turn, lead to larger domain sizes. Thus, we exposed the LC/monomer mixture using a high UV intensity (I~45 mW/cm²). The exposure time is 30 min. The resultant polymer networks align, on the average, parallel to the substrate surfaces due to the strong surface anchoring energy. Thus, the PNLC cell is highly transparent in the voltage-off state. In a voltage-on state, the polymer networks exert a torque to resist the LC molecules from being reoriented by the electric field. As a result, the threshold voltage is increased significantly. Once the electric field is removed, the strong polymer field assists LC to relax back quickly.

For laser beam steering at λ=1.55 μm, both reflective and transmissive phase modulators can be used. To generate diffractive phase gratings, a 2π phase change between adjacent grating elements is required. The reflective and transmissive PNLC cells have been prepared and investigated.

Figure 1 depicts the experimental setup for studying the electro-optic effects of the reflective and transmissive PNLC cells. The incoming diode laser beam (λ=1.55 μm) is reflected or transmitted by the polarizing beam splitter (PBS) to the PNLC cell. The rubbing direction of the LC cell is oriented to be 45° to the polarization axis of the incident beam. For the reflective mode, ideally the reflector should be imbedded in the inner side of the rear substrate in order to eliminate multiple surface reflections. For feasibility studies, we placed a dielectric mirror behind the LC cell to serve as a reflector. For the transmissive mode, the analyzer is crossed to the polarizer. The voltage-dependent reflectance or transmittance of the cells is recorded by the LabVIEW system and data are analyzed by a computer.
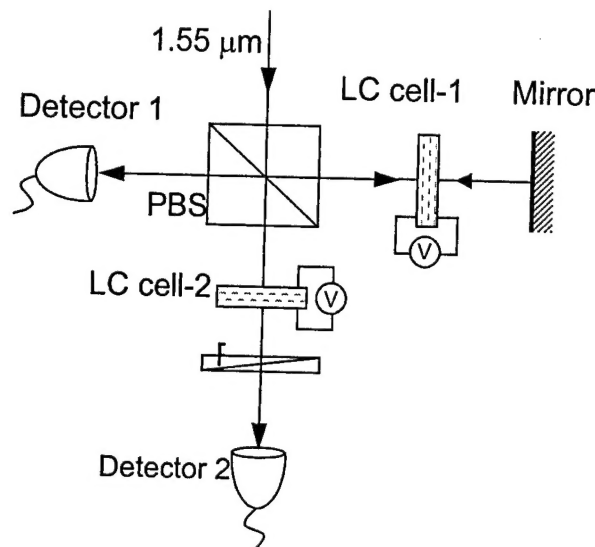
Figure 1. Experimental setup for measuring the electro-optic properties of the reflective and transmissive LC and PNLC cells. The rubbing direction of the cell is at 45° to the polarization axis of the incident light. The LC cell-1 and detector 1 are for reflective mode and LC cell-2 and detector 2 are for transmissive mode. PBS = polarizing beam splitter. P = Polarizer. $\lambda = 1.55$ μm.

Two types of diacrylate monomers: bisphenol-A-dimethacrylate (M1) and RM-82 (Merck) were used. They have a rod-like structure. The melting point of these two monomers is 74.6 and 82.4 °C, respectively. Due to the high heat fusion enthalpy, each monomer's solubility is limited to 7-8 wt%. Figure 2 shows the voltage-dependant reflectance of PNLC cells with different monomer concentrations. When monomer concentration is 7 wt%, the cured polymer network domain size is still too large which results in ~10% light scattering loss at λ=1.55 μm. In order to eliminate light scattering, we need to reduce the domain size further by increasing the monomer concentration. Therefore, we mixed M1 with RM-82 to lower the melting point. For the LC mixture (Merck E44) we studied, the best monomer concentration is 6 wt% M1 and 4 wt% RM-82, shown as the black solid line in Fig. 2. Unless otherwise mentioned throughout this paper, our LC/polymer contains 90% LC, 6% M1 and 4% RM-82.
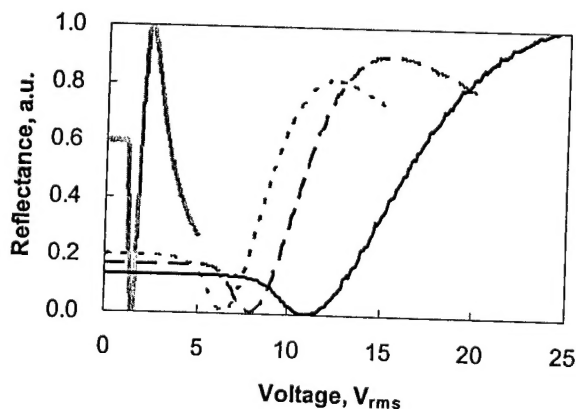

Figure 2. Voltage-dependent reflectance of E44 (gray lines) and PNLC (dark lines) cells. From left to right, c=5, 7, and 10 wt%. The cell gap for the reflective cells is 5 μm. λ=1.55 μm and T=21 °C.

3.2.3 Results and Discussion

Figures 3 (a) and (b) plot the voltage-dependent reflectance and transmittance of the corresponding 7.7 μm and 12.5 μm E44 PNLC cells. The total phase retardation of the reflective and transmissive PNLC cells is measured to be δ=3.84π and 3.0π, respectively. However, the last π phase change would require a very large voltage swing. For the

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interest of keeping operating voltage low, we operated the LC modulator from $\delta = 2\pi$ (where $V = V_{2\pi}$) to 0 (V=0). Also included in Figs. 3 (a) and (b) (gray lines) for comparisons are the 7.7-μm and 12.5-μm E44 cells without polymer. The threshold voltage of the E44 cells is ~0.9 V_{rms}. From the measured total phase retardation, the birefringence of E44 at λ=1.55 μm is Δn~0.21 at T~21 °C. On the other hand, the Δn of the PNLC cell was measured to be Δn~0.19. The lower Δn originates from the imbedded 10% polymers. This result indicates that the polymer networks do not disturb LC alignment noticeably. The threshold voltage of the 7.7-μm and 12.5-μm PNLC cells is increased to 20 and ~24 V_{rms}, respectively. The increased threshold voltage implies that the LC molecules are tightly anchored by the polymer networks. In Figs. 3 (a) and (b), the peak transmittance of the PNLC cells is similar to that of the pure E44 cells. This indicates that the light scattering is negligible. We also measured the wavelength-dependent transmittance of the 12.5 μm cell. Scattering is strong in the visible region but becomes negligible at λ>1μm.
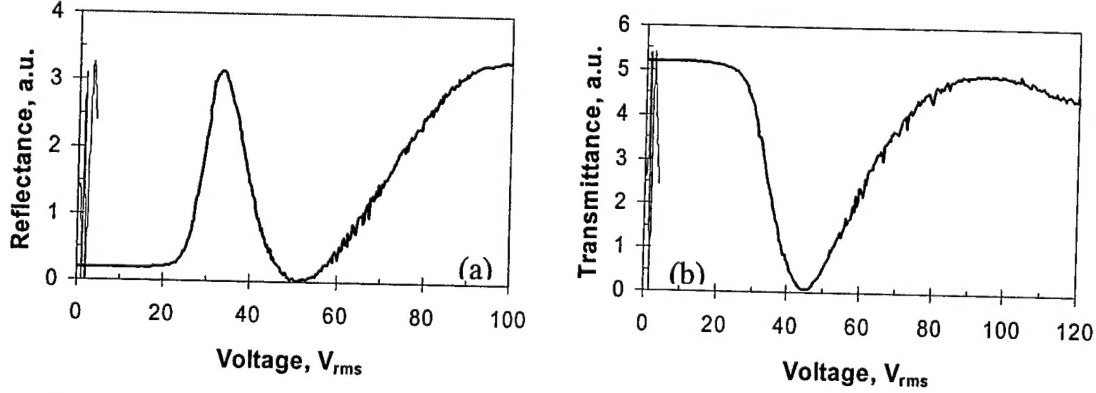

Figure 3. Voltage-dependent reflectance (a) and transmittance (b) of E44 (gray lines) and PNLC (dark lines) cells. The cell gap for the reflective and transmissive cells is 7.7 μm and 12.5 μm, respectively. λ=1.55 μm and T=21 °C.

Figures 4(a) and (b) show the measured decay time (solid lines) of the reflective and transmissive PNLC cells. The open circles are fittings with exponential phase decay: $\delta(t) = 2\pi \exp(-t/\tau)$. Good agreement is found. The time constant (at 1/e) for the 7.7-μm reflective cell and 12.5-μm transmissive cell is τ=296 μs and 755 μs, respectively. If we count the phase decay time from 100→10%, they are ~0.7 ms and 1.8 ms, respectively. As compared to the pure E44 cell, the response time is improved by ~300X. The rise time (10→90% phase change) was measured to be ~0.5 ms for the transmissive PNLC cell operating from 0 to 90 V_{rms}.
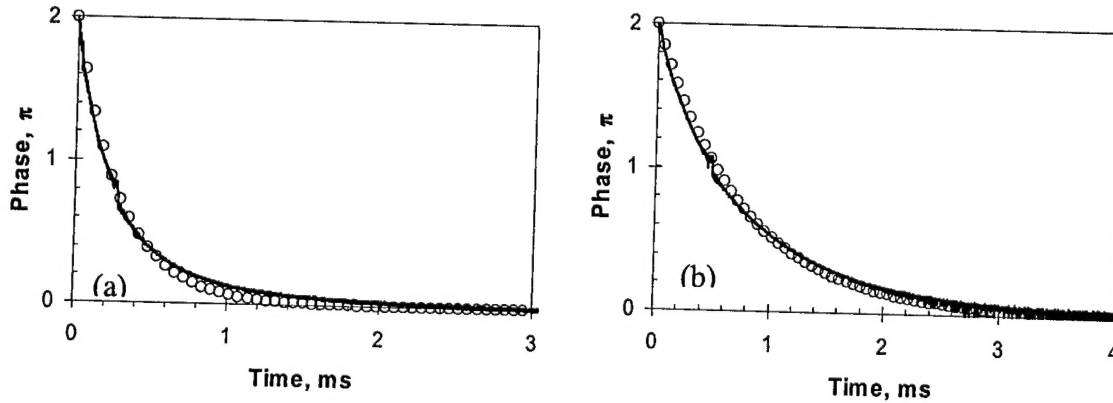

Figure 4. Measured phase decay time of the reflective (a) and transmissive (b) PNLC cells. Solid lines are experimental data and open circles are fitting results. In (a), τ=296 μs and in (b) τ=755 μs. T=21°C and λ=1.55 μm.

Figures 5 (a) and (b) show the simulation results of the pure E44 and PNLC cells. For the E44 cell, in Fig. 5 (a), the following measured parameters are used: Δε=15.5, K_{11}=15.3 pN, and γ_1=0.448 Pas. The ratio of γ_1/K_{11} is 29.33 ms/μm². We assumed the pretilt angle is 2.5°. From fitting, we find K_{33}=24.2 pN. For the PNLC cell, we assume the Δε and γ_1 of E44 do not change because the pure LC molecules are completely separated from the polymer. In the

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PNLC cell, the measured γ_1/K_{11} drops to 0.094 ms/μm^2, indicating a much faster decay time. The expected effective K_{11} of PNLC cell would be 4773 pN. From the fittings shown in Fig. 5 (b), we find K_{11}=4800 pN and K_{33}=50000 pN. The simulation results imply that the strong anchoring force of polymer network in bulk LC area dramatically increases the effective K value. The LC molecules in the polymer network domains become stiffer and more difficult to be reoriented by the electric field. A small portion of the bulk LC molecules near the boundaries of the network domain may not rotate at all, which results a lower phase change in the high voltage regime.

Figure 6 shows the temperature dependent birefringence of the transmissive E44 PNLC cell at λ=1.55 μm. The dots represent the experimental data and solid lines are fittings with Haller's equation $\Delta n = \Delta n_o (1 - T/T_c)^{\beta}$; where Δn_o is the birefringence at T=0 and β is a material parameter. From the fittings, we find Δn_o =0.255 and β=0.187.

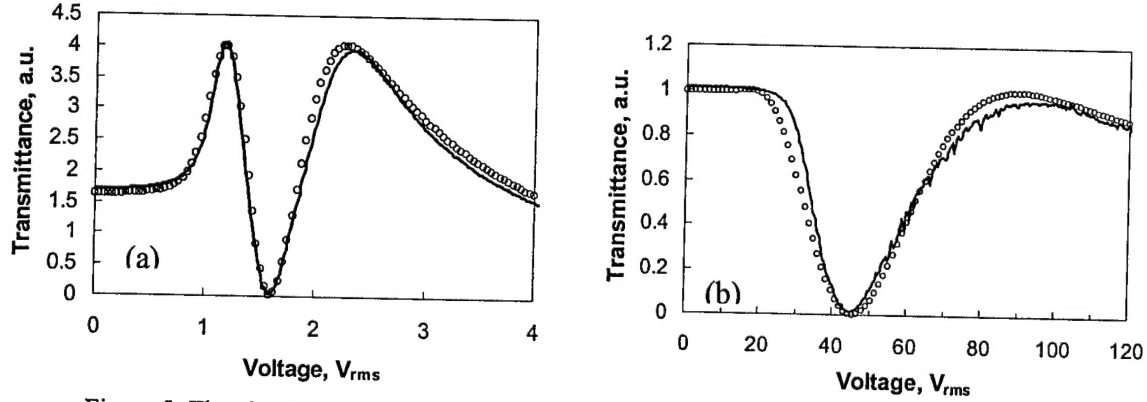Figure 5. The simulation results of E44 (a) and PNLC (b) cells. Solid lines are experimental data and open circles are fitting results. T=21°C and λ=1.55 μm.
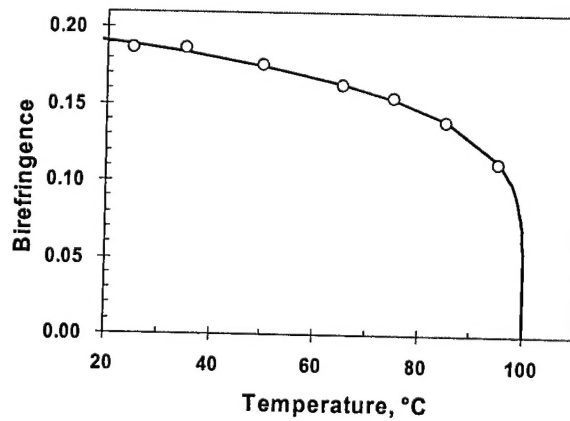

Figure 6. Temperature-dependent birefringence of the E44 transmissive PNLC cell. d=12.58 μm.

To reduce the operating voltage, we cured the PNLC cell under a bias voltage V_b. The 8-μm PNLC cells were prepared at V_b=0, 0.9, 1.0, and 1.1 V_{rms}, as shown in Figure 7. The high threshold voltage is reduced when V_b=0.9 V_{rms} and eliminated when V > 1.0 V_{rms}. The tradeoff is that the response time is slightly increased. The bias voltage needs to be optimized in order to maintain the maximum phase change and fast response time.
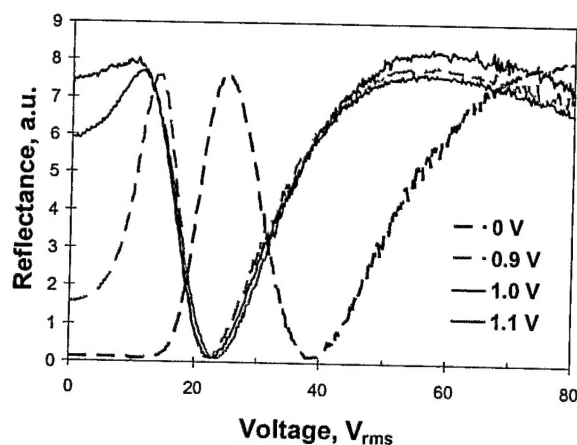
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Figure 7. Voltage-dependent reflectance PNLC cells cured with different bias voltage. The cell gap for the reflective cells is 8 μm. λ=1.55 μm and T=21 °C.

To reduce the operating voltage while retaining the required 2π phase change, we could increase the LC birefringence and dielectric anisotropy ($\Delta\varepsilon$). Figure 8 shows the simulation results of the PNLC cell as Δn and $\Delta\varepsilon$ increase. The $V_{2\pi}$ could be reduced to 54.5 V if Δn is increased to 0.3, as shown in Fig. 8 (a). We could lower both V_{th} and $V_{2\pi}$ by using a higher $\Delta\varepsilon$ material. The $V_{2\pi}$ is decreased gradually as $\Delta\varepsilon$ increases, as shown in Fig. 8 (b).

Our experimental data show that the response time of the PNLC cell is linearly proportional to the cell gap. Thus, for practical applications a proper balance between operating voltage and response time needs to be taken into consideration.

The developed PNLC can also be extended to the 3-5 and 8-12 μm spectral bands. In the long wavelength region, the LC birefringence decreases but the light scattering becomes less significant. To obtain 2π phase change, we need a thicker LC layer. The increased cell gap would lead to a higher voltage. To reduce voltage, we could increase the domain size.
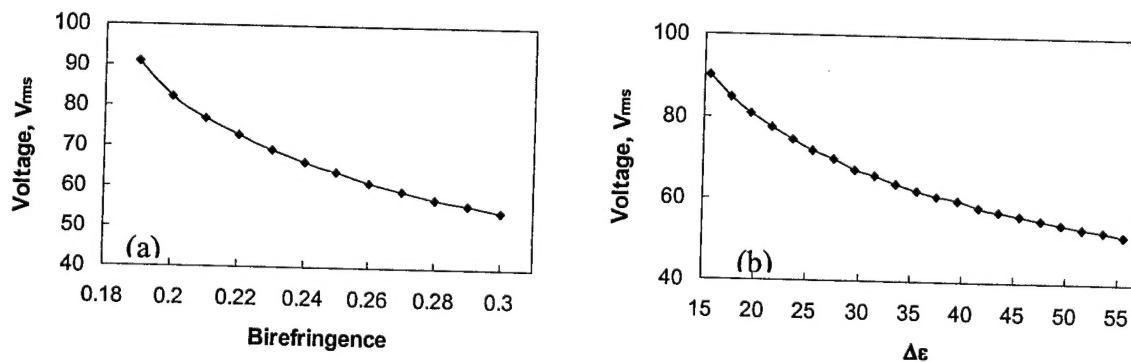

Figure 8. The simulation results of PNLC cell with different Δn (a) and $\Delta\varepsilon$ (b). The fitting parameters: K_{11}=4800 pN, and K_{33}=50000 pN. λ=1.55 μm.

We have demonstrated fast-response and scattering-free reflective and transmissive PNLC phase modulators. The shortcoming is the increased voltage. To reduce operating voltage, high Δn and large $\Delta\varepsilon$ LC mixtures need to be developed. The PNLC cell also holds promise for mid and long infrared applications.

4. Personnel Supported
One post doctorate and 2 graduate students.

5. Publications: (9/2003 to 8/2004)

1. F. Du, S. Gauza and S. T. Wu
    "Influence of curing temperature and high birefringence on the properties of polymer-stabilized liquid crystals", Optics Express 11, 2891-6 (Nov. 3, 2003).

2. Y. H. Fan, H. W. Ren and S. T. Wu
    "Switchable Fresnel lens using polymer-stabilized liquid crystals,"
    Optics Express 11, 3080-6 (Nov. 17, 2003).

3. Y. Q. Lu, F. Du And S. T. Wu
    "Polarization Switch Using Thick Holographic Polymer-Dispersed Liquid Crystal Grating",
    J. Appl. Phys. 95, 810-5 (Feb. 1, 2004)

4. J. Li and S. T. Wu
    "Extended Cauchy Equations For The Refractive Indices Of Liquid Crystals",
    J. Appl. Phys. 95, 896-901 (Feb. 1, 2004)

5. Y. H. Fan, Y. H. Lin, H. Ren, S. Gauza, and S. T. Wu
    "Fast-Response and Scattering-Free Polymer Network Liquid Crystals"
    Appl. Phys. Lett. 84, 1233-5 (Feb. 23, 2004)

6. Y. Huang, S. T. Wu, and Y. Zhao
    "All-Optical Switching Characteristics in Bacteriorhodopsin And Its Applications In Integrated Optics"
    Optics Express 12, 895-906 (March 8, 2004)

7. Y. Lu, C. Wong And S. T. Wu "Liquid Crystal-Based Fourier Optical Spectrum Analyzer" IEEE Photonics Technology Letters 16, 861-3 (March, 2004)

8. Y. Q. Lu, F. Du, Y. H. Lin, and S. T. Wu
    "Variable Optical Attenuator Based On Polymer-Stabilized Twisted Nematic Liquid Crystal"
    Optics Express 12, 1221-7 (April 5, 2004).

9. Q. H. Wang, T. X. Wu, X. Y. Zhu, and S. T. Wu
    "Achromatic Polarization Switch Using a Twisted Nematic Liquid Crystal"
    Liq. Cryst. 31, 535-9 (April, 2004)

10. C. O. Catanescu, S. T. Wu, and L. C. Chien
    "Tailoring the Physical Properties of Some High Birefringence Isothiocyanato-Based Liquid Crystals"
    Liq. Cryst. 31, 541-555 (April, 2004)

11. J. Li, S. Gauza, and S. T. Wu
    "High Temperature-Gradient Refractive Index Liquid Crystals"
    Optics Express 12, 2002-10 (May 3, 2004).

12. Y. H. Lin, H. W. Ren, and S. T. Wu
    "High Contrast Polymer-Dispersed Liquid Crystal in a 90° Twisted Cell"
    Appl. Phys. Lett. 84, 4083-5 (May 17, 2004).

13. P. T. Lin, S. T. Wu, C. Y. Chang, and C. S. Hsu
    "UV Stability of High Birefringence Liquid Crystals"
    Mol. Cryst. Liq. Cryst. 411, 243-253 (May 2004)

14. Carmen O. Catanescu, L. C. Chien, and S. T. Wu
    High Birefringence Nematic Liquid Crystals for Display Applications
    Mol. Cryst. Liq. Cryst. 411, 93-102 (May, 2004)

15. H. Wang, T. X. Wu, X. Zhu and S. T. Wu
    "Correlations Between Liquid Crystal Director Reorientation and Optical Response Time Of A Homeotropic Cell",
    J. Appl. Phys. 95, 5502-8 (May 15, 2004)

16. X. Zhu And S. T. Wu
    "Normally Black Reflective Twisted Nematic Cell for Microdisplay Application"
    J. Appl. Phys. 95, 7660-4 (June 15, 2004).

17. Y. B. Huang, H. P. Shieh and S. T. Wu
    "Applications of Multidirectional Asymmetrical Microlens Array Light Control Films On Reflective Liquid Crystal Displays For Image Quality Enhancement"
    Applied Optics 43, 3656-63 (June 20, 2004).

18. J. Li and S. T. Wu
    "Temperature Effect on the Liquid Crystal Refractive Indices"
    J. Appl. Phys. 96, 19-24 (July 1, 2004).

19. J. Li and S. T. Wu
    "Two-Coefficient Cauchy Model for Low Birefringence Liquid Crystals"

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J. Appl. Phys. 96, 170-4 (July 1, 2004).
20. P. T. Lin, X. Liang, H. Ren, and S. T. Wu
"Tunable Diffraction Grating Using UV-Induced Spatial Phase Modulation in Dual-Frequency Liquid Crystal"
Appl. Phys. Lett. 85, 1131-1133 (Aug. 16, 2004).
21. F. Du, Y. Q. Lu, and S. T. Wu
"Electrically Tunable Liquid Crystal Photonic Crystal Fiber"
Appl. Phys. Lett. (Sept. 20, 2004).
22. Y. H. Fan, H. Ren, Y. H. Lin, and S. T. Wu
"Dual Frequency Liquid Crystal Gels With Sub-Millisecond Response"
Appl. Phys. Lett. (Sept. 27, 2004).
23. Y. Q. Lu, X. Liang, Y. H. Wu, F. Du, and S. T. Wu
"Dual-Frequency Addressed Hybrid-Aligned Liquid Crystal"
Appl. Phys. Lett. (Oct. 25, 2004).

6. Interactions or Transitions: (2003-2004 only)

A. Presentations:
1. S. T. Wu
"Molecular Engineering of High Birefringence Liquid Crystals"
(Keynote speaker) 15th Conference on Liquid Crystal (Zakopane, Poland, Oct. 13-17, 2003)
2. S. T. Wu
"Tunable-focus liquid crystal lens"
(Invited speech) Department of Material Science and Engineering, University of Florida (Dec. 9, 2003)
3. S. T. Wu
"Liquid Crystal Device Physics"
(One-week Display Winter School) National Chiao Tung University, Hsinchu, Taiwan (Feb. 2-6, 2004)
4. S. T. Wu
"Liquid Crystal Displays and Foveated Imaging"
(Keynote Speaker) School of Optics Industrial Affiliates Day (April 16, 2004)
5. S. T. Wu
"Photostability of LCoS devices"
(Invited talk) eLCOS Microdisplays (Sunnyvale, CA, May 20, 2004).
6. S. T. Wu
"Accelerated life test of liquid crystals and alignment layers"
(Invited talk) Intel Corp. Santa Clara, California (May 20, 2004)
7. S. T. Wu
"High contrast PDLC for reflective displays"
(Invited talk) SiPix Imaging, Miltipas, California (May 21, 2004)
8. S. T. Wu
"Reflective and transflective LCDs"
SID annual meeting, Seminar M-8, Seattle, WA (May 24, 2004) (~200 audiences)

B. Transitions:

1. We shipped a high birefringence liquid crystal mixture to Dr. Terry Dorschner of Raytheon for testing his laser beam steering. Telephone number: 978-684-8738.

2. We shipped a high birefringence liquid crystal mixture to Dr. Teresa Ewing of Boulder Nonlinear Systems, for testing her mid-infrared dynamic scene projector. Telephone number: 303-604-0077.

3. We provided a high birefringence dual-frequency LC sample to Dr. Dong-Feng Gu of Rockwell Scientific Company for testing his optical phased arrays. Email: dgu@rwsc.com.

7. Honors and Awards

S. T. Wu is a Fellow of the IEEE, Society for Information Display (SID), and Optical Society of America (OSA). He received the IEEE Outstanding Engineer award in 2003, SID special recognition award in 2000, and SID Distinguished Student paper award (2004). He has published two books: 1. "Optics and Nonlinear Optics of Liquid Crystals" (World Scientific, Singapore, 1993; with Prof. I. C. Khoo) and 2. "Reflective Liquid Crystal Displays" (Wiley-SID, 2001; with Prof. D. K. Yang). In 2004, he was elected to the founding editor-in-chief of the IEEE/OSA Journal of Display Technology.